



Cesium carbonate-doped 1,4,5,8-naphthalene-tetracarboxylic-dianhydride used as efficient electron transport material in polymer solar cells

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ABSTRACT

The use of appropriate charge carrier transport materials in organic solar cells strongly influences the device performance. In this work, we focused on the molecular electron transport material 1,4,5,8-naphthalene-tetracarboxylic-dianhydride (NTCDA) doped by cesium carbonate (Cs_2CO_3). We first investigated the electrical properties of such n-type doped material as a function of the doping concentration before using it as electron transport layer (ETL) in polymer solar cells. The doped transparent ETL reduces the series resistance leading to an increased open circuit voltage. A power conversion efficiency of 3.8% was finally achieved in a device with a blend of poly(3-hexylthiophene-2,5-diyl):phenyl-C61-butyric acid methyl ester (P3HT:PCBM) as the active layer and a 5 nm-thick NTCDA: Cs_2CO_3 film with a molar ratio of 30% as ETL.

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1. Introduction

The multitude of research efforts in organic photovoltaic cells have led to the achievement of a power conversion efficiency of 12% on an active surface of 1.1 cm^2 by Heliotech in 2013 [1]. The development of these potentially low-weight, flexible and semi-transparent solar cells, which could additionally involve less expensive and less pollutant production processes than those of inorganic solar cells, is the main objective of many research efforts. Both efficiency and lifetime need to be improved and this could be achieved by engineering the interfaces between the electrodes and the organic active layer.

Extraction and transport of charge carriers are crucial in organic optoelectronic devices and therefore, sufficiently conductive and stable transport layers are required in order to reduce ohmic losses. Doping organic semiconductors represents one of the possible approaches to optimize

these interfaces. It increases the density of free carriers leading to an enhanced electrical conductivity. P-type doping can easily be created by incorporating a strong electron acceptor molecule like the widely investigated 2,3,5,6-Tetrafluoro-7,7,8,8-tetracyanoquinodimethane ($\text{F}_4\text{-TCNQ}$) in matrices such as Zinc Phthalocyanine (ZnPc) [2,3] or 4,4',4''-tris(3-methyl-phenyl-phenyl-amino)triphenylamine (m-MTDATA) [4]. However n-type dopants need to be strong electron donors and are consequently rare because of stability issues due to oxidation. Alkali metals [5] and cationic dyes [6,7] have been successfully used as n-type dopants. Cesium carbonate (Cs_2CO_3) can also be used as a suitable n-type dopant and has already been combined with organic semiconductors such as fullerene C60 [8,9] or 4,7-diphenyl-1,10-phenanthroline (Bphen) [10]. Additionally, it has been demonstrated that Cs_2CO_3 could reduce the work function of the cathode and improves charge injection allowing the fabrication of efficient both conventional and inverted polymer solar cells [11–15].

Here we report an efficient n-type doping of the transparent electron transport material 1,4,5,8-naphthalene-

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tetracarboxylic-dianhydride (NTCDA) with Cs_2CO_3 . First, the electrical and morphological properties of NTCDA: Cs_2CO_3 thin films were analyzed as a function of the dopant concentration. Afterwards, knowing that co-sublimated doped charge transport layers have been abundantly used for organic electronic applications such as organic light emitting diodes (OLED) and small molecule organic solar [16–19] cells, but not extensively in polymer photovoltaic devices [20,21], we investigated the effect of the incorporation of an n-doped NTCDA layer in poly (3-hexylthiophene) (P3HT):phenyl-C61-butyric acid methyl ester (PCBM) bulk heterojunction solar cells. Indeed, this type of device conventionally includes a metal oxide such as zinc oxide [22,23] or titanium oxide [24,25] as electron transport layer.

2. Experimental details

2.1. Co-sublimation and characterization of Cs_2CO_3 -doped NTCDA thin films

Cs_2CO_3 -doped NTCDA thin films were deposited by co-sublimating at constant rates both materials on glass substrates in a vacuum chamber at 10^{-6} mbar. Evaporation rates and consequently molar ratios were controlled by using quartz microbalances with a mass density of 1.2 and 4 for NTCDA and Cs_2CO_3 respectively. These films were then characterized. First, the electrical conductivity was determined by using a four-point probe under 10^{-2} mbar as a function of the temperature. The Seebeck coefficient was measured with two heating points connected to thermocouples and an electrometer (Keithley 617). An atomic force microscope (CSI Nano-observer) was used to produce AFM images of the NTCDA: Cs_2CO_3 thin films.

2.2. Fabrication and characterization of polymer solar cells

Organic solar cells were built on commercial ITO-coated glass substrates with a sheet resistance of 7Ω per square. First of all, the substrate was cleaned by sonication in acetone, ethanol and isopropyl alcohol. A thin layer of Cs_2CO_3 -doped NTCDA was then co-sublimated at 10^{-6} mbar on the ITO glass substrate and annealed at 150°C for 30 min

in a nitrogen-filled glove box. Annealing is known to remove water from the layer [26]. Furthermore, it was observed that annealed Cs_2CO_3 can be used as ETL in polymer solar cell on the ITO electrode. This annealing step reduces the work function of the transparent ITO electrode and therefore improves charge extraction [27,28]. Afterwards, a 180 nm-thick layer of P3HT:PCBM was deposited by spin coating a 54 mg mL^{-1} solution of P3HT:PCBM (1:0.8 by weight in *o*-dichlorobenzene) at 1200 rpm for 40 s. Immediately after the deposition of the active layer, each sample was placed for 15 min in a closed Petri dish for a slow drying and heated at 50°C for 15 min to complete the evaporation of the solvent. A thin layer of poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT-PSS) was then deposited by spin coating the Clevis F010 solution (as provided) on the active layer at 5500 rpm for 50 s. Finally, an 80 nm-thick electrode of silver was evaporated at 10^{-6} mbar and the device was annealed at 110°C for 15 min on a hot plate. A final device consists of 2 rectangular cells of 18 mm^2 as illustrated in Fig. 1a. Current–voltage characteristics of the photovoltaic cells were measured at room temperature under simulated AM1.5G solar illumination of 100 W/cm^2 with a Keithley 2400 source meter, taking into account the spectral mismatch of the sun simulator.

3. Results and discussion

3.1. Dopant concentration and temperature dependence

As highlighted in Fig. 2, the dopant Cs_2CO_3 presents a highest occupied molecular orbital (HOMO) close to the lowest unoccupied molecular orbital (LUMO) of the NTCDA which enables an electron transfer to NTCDA increasing consequently the charge carrier concentration and the electrical conductivity. Energy level values are taken from the literature [11,19,29]. As shown in Fig. 3a, the electrical conductivity measured at room temperature increases with the Cs_2CO_3 concentration and reaches a maximum around 10^{-3} S/cm for a molar ratio $N_{\text{Cs}_2\text{CO}_3}/N_{\text{NTCDA}}$ around 50% which is 5 orders of magnitude higher than the conductivity of undoped NTCDA [30,6]. For high dopant concentrations, the AFM images of Fig. 4 show that cesium carbonate aggregates and causes structural changes in-

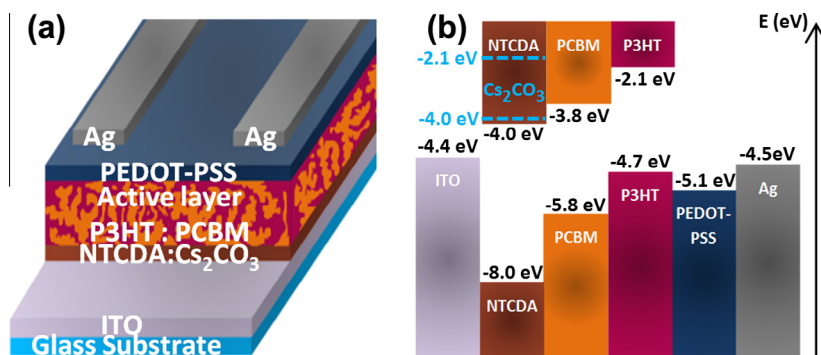


Fig. 1. Device architecture (a) and energy band diagram (b) of P3HT:PCBM-based solar cell including a Cs_2CO_3 -doped NTCDA electron transport layer. Work functions, ionization energies and electronic affinity are typical indicative values found in the literature [40–42,19]. The blue dashed horizontal lines at 4 eV and 2.1 eV on the NTCDA diagram represent the HOMO and the LUMO of the dopant, respectively [11]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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